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Review

Polychlorinated Biphenyls (PCBs) in Poland: Occurrence, Determination and Degradation

Iwona Grabowska*

Institute of Animal Reproduction and Food Research, Polish Academy of Sciences,
Tuwima 10, 10-747 Olsztyn, Poland

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Abstract

One of the main global ecological problems is contamination of the natural environment by persistent organic pollutants (POPs). Polichlorinated biphenyls (PCBs) is a group of compounds that belongs to this family of pollutants.

Poland, as a European Union member and signatory of international agreements concerning environmental protection, is obliged to regulate the law and intensify work on the control and utilization of biphenyl PCBs.

Therefore, the occurence and determination of PCBs in the polish environment (rivers, lakes, sea water and sediments, living organisms and food chains), and the toxicity and degradation methods of these POP compounds is still important.

Keywords: polichlorinated biphenyls (PCBs), organic pollutants, evironment

Introduction

Polychlorinated biphenyls (PCBs) belong to a class of organic compounds containing 1-10 atoms of chlorine attached to biphenyl, by chemical formula $C_{12}H_{10-x}Cl_x$, where $x \geq 1$. The general chemical structure of this class of compounds is presented in Fig. 1.

The synthesis of PCBs is based on direct chlorination of biphenyl by catalysis of $FeCl_3$. The reaction goes through the free radicals mechanism, so the products contain a mixture of biphenyls with different numbers of chlorine atoms. Separation of these compounds seems to be impossible. Because of that, mixtures of PCBs were available in various countries, under various comercial brand names, such as “Aroclor” (USA), “Clophen” (Germany), “Phenoclor” (France), “Fenchlor” (Italy), “Kanechlor” (Japan), “Tarnol” and “Chlorofen” (Poland) [1, 2].

*e-mail: i.grabowska@pan.olsztyn.pl

There are possibilty 42 isomers of tetrachlorobiphenyls and hexachlorobiphenyls. Overall, there are 209 congeners of PCBs, strarting from chlorobiphenyl to completely substituted dekachlorobiphenyl (Table 1). The nomenclature of PCB homologues according to IUPAC is presented in Table 2.

Historically, PCBs were first produced in 1929 and used as components of paints, especially printings. Then these compounds were used as plastificator of plastics and insulating materials in transformers and capacitors, heat transfer

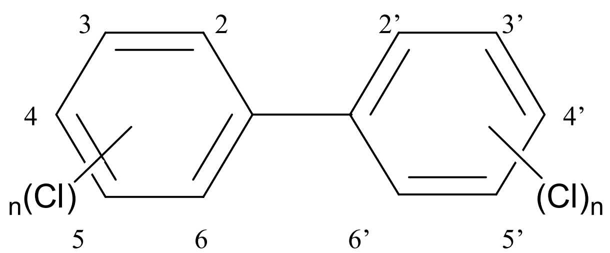

Fig. 1. Chemical structure and numbering of PCB.

Table 1. Homologous series of chloroderivatives of biphenyl.

Homologue	Molecular formula	Number of isomers	IUPAC No.
Monochlorobiphenyl	$C_{12}H_9Cl$	3	1 - 3
Dichlorobiphenyl	$C_{12}H_8Cl_2$	12	4 - 15
Trichlorobiphenyl	$C_{12}H_7Cl_3$	24	16 - 39
Tetrachlorobiphenyl	$C_{12}H_6Cl_4$	42	40 - 81
Pentachlorobiphenyl	$C_{12}H_5Cl_5$	46	82 - 127
Hexachlorobiphenyl	$C_{12}H_4Cl_6$	42	128 - 169
Heptachlorobiphenyl	$C_{12}H_3Cl_7$	24	170 - 193
Oktachlorobiphenyl	$C_{12}H_2Cl_8$	12	194 - 205
Nonachlorobiphenyl	$C_{12}HCl_9$	3	206 - 208
Decachlorobiphenyl	$C_{12}Cl_{10}$	1	209

fluids, additives in hydraulic fluids in vacuum and turbine pumps [1, 2], because of such properties as: low dielectric constant, low rates of evaporation, chemical intertness, non-flammability, high heat capacity, and high electrical resistivity. Since the 1970s, production of these compounds has been limited or even terminated in many countries. So far, 2/3 of the total amount of PCBs produced globally (1.5 tons) is still used or present in the environment.

It has been known since 1966 that these compounds are harmful. They are classified as persistent organic pollution (POP) and are spread throughout the environment. Concentrations of PCBs in the environment and organisms is presented in Table 3. The main reasons for the presence of these compounds in the environment could be as follows [3]:

- leakage from capacitors and transformers,
- uncontrolled storage of industrial waste oils (transformer, capacitor, motor, etc.),
- improper regeneration of transformer and motor oil,
- inappropriate disposal practices.

The purpose of this review is to present the results of research on PCB occurrence in Poland, their toxicity, methods of determination and degradation. The data published prove that these compounds are present in the water of Polish river ecosystems, in bottom sediments, and in living organisms such as pikerperch, red deer, freshwater mussels. PCBs also are found in Polish milk and diairy products, but at acceptable levels.

Among the methods of PCB determination, conventional analytical methods such as gas chromatogrpahy (GC) and high performance liquid chromatography (HPLC) are frequenly used. However, the immunochemical or potentiometric methods of determination cannot be ommited. Moreover, their neurotoxic influence on humans and animals is also very well documented. Very important from a practical point of view is the degradation of PCBs, e.g. by chemicall processes or microbiological degradation.

This review includes all these aspects connected with PCBs in Poland, and a comparison with problems presented in the scientific literature.

The problem seems to be current. Council Directive 96/59/EC of 16 September 1996 on the disposal of polychlorinated biphenyls and polychlorinated terphenyls (PCB/PCT) states that equipment and PCBs contained in inventories must be decontaminated or disposed of by 2010 in European Union member countries [4]. On the other hand, the Stockholm Convention on Persistent Organic Pollutants has imposed the elimination of the use of PCBs in equipment by 2025, and the disposal of PCBs by 2028 [5].

Occurrence of PCB in:

Water Reservoirs, Bottom Sediments and Sludge

In the scientific literature, the influence of the following parameters on the concentration and movement of PCBs in an aqueous environment were studied: historical and current industrial facilities [6, 7], seasonal trends and upstream tributaries [6], floods [7], climatic regime and settling fluxes [8]. Because of hydrophobic properties and, consequently, low solublity in water, PCBs are more strongly accumulated in sediments and biota [6-8].

The occurrence of PCBs in water reservoirs and bottom sediment samples has been widely studied in Poland (e.g. within the framework of the International Odra Project) [9-11].

A research group from Gdańsk University of Technology was engaged in this project, indicating that water from the Odra River and its tributaries and sediments are not polluted with PCBs at unacceptable levels. They conducted measurements on samples collected between 1997-2000, among others immediately after the flood in 1997 [9, 10]. Also, another group of researchers, from the Department of Water Pollution Control in Gdańsk, determined the levels of PCBs in sediments from different sites of the Odra and its tributaries during 1998-2002 [11]. They found that concentrations of PCBs varied significantly, and that the highest content of PCBs 180 and 138 were present in sediments investigated from the Odra in the region between the Nysa Łużycka confluence and Mescherin. Authors showed the influence of the 1997 flood, and on the increase of PCB concentrations in the studied sediments of the Odra and its tributaries [11].

The other research on the determination of concentration profiles of PCBs were done based on sediment samples taken in 1992 from the Vistula River and Gdańsk Depth in the Baltic Sea, and samples collected in 1992 from a sewage treatment plant in Dębogórze (city of Gdynia) [12]. Among organochlorine compounds analyzed, PCBs dominated in sludge samples as described by these authors [12]. On the other hand, a group from Poznań University of Medical Sciences determined succesfully 15 selected PCBs in sediments using the fexIKA extraction method [13].

Table 2. Nomenclature of PCB – homologues according to IUPAC.

No.	Name	No.	Name	No.	Name	No.	Name	No.	Name
MonochloroCB		42	2,2',3,4'	85	2,2',3,4,4'	**HexachloroCB**		171	2,2',3,3',4,4',6
1	2	43	2,2',3,5	86	2,2',3,4,5	128	2,2',3,3',4,4'	172	2,2',3,3',4,5,5'
2	3	44	2,2,3,5'	87	2,2',3,4,5'	129	2,2',3,3',4,5	173	2,2',3,3',4,5,6
3	4	45	2,2',3,6	88	2,2',3,4,6	130	2,2',3,3',4,5'	174	2,2',3,3',4,5,6'
DichloroCB		46	2,2',3,6'	89	2,2',3,4,6'	131	2,2',3,3',4,6	175	2,2',3,3',4,5',6
4	2,2'	47	2,2',4,4'	90	2,2',3,4',5	132	2,2',3,3',4,6'	176	2,2',3,3',4,6,6'
5	2,3	48	2,2',4,5	91	2,2',3,4',6	133	2,2',3,3',5,5'	177	2,2',3,3',4',5,6
6	2,3'	49	2,2',4,5'	92	2,2',3,5,5'	134	2,2',3,3',5,6	178	2,2',3,3',5,5',6
7	2,4	50	2,2',4,6	93	2,2',3,5,6	135	2,2',3,3',5,6'	179	2,2',3,3',5,6,6'
8	2,4'	51	2,2',4,6'	94	2,2',3,5,6'	136	2,2',3,3',6,6'	180	2,2',3,4,4',5,5'
9	2,5	**TetrachloroCB**		95	2,2',3,5',6	137	2,2',3,4,4',5	181	2,2',3,4,4',5,6
10	2,6	52	2,2',5,5'	96	2,2',3,6,6'	138	2,2,3,4,4',5'	182	2,2',3,4,4',5,6'
11	3,3'	53	2,2',5,6'	97	2,2',3',4,5	139	2,2',3,4,4',6	183	2,2',3,4,4',5',6
12	3,4	54	2,2',6,6'	98	2,2',3,4,6	140	2,2',3,4,4',6'	184	2,2',3,4,4',6,6'
13	3,4'	55	2,3,3',4	99	2,2',4,4',5	141	2,2',3,4,5,5'	185	2,2',3,4,5,5',6
14	3,5	56	2,3,3',4'	100	2,2',4,4',6	142	2,2',3,4,5,6	186	2,2',3,4,5,6,6'
15	4,4'	57	2,3,3',5	101	2,2',4,5,5'	143	2,2',3,4,5,6'	187	2,2',3,4',5,5',6
TrichloroCB		58	2,3,3',5'	102	2,2',4,5,6'	144	2,2',3,4,5',6	188	2,2',3,4',5,6,6'
16	2,2',3	59	2,3,3',6	103	2,2',4,5',6	145	2,2',3,4,6,6'	189	2,3,3',4,4',5,5'
17	2,2',4	60	2,3,4,4'	104	2,2',4,6,6'	146	2,2',3,4',5,5'	190	2,3,3',4,4',5,6
18	2,2',5	61	2,3,4,5	105	2,3,3',4,4'	147	2,2',3,4',5,6	191	2,3,3',4,4',5',6
19	2,2’,6	62	2,3,4,6	106	2,3,3',4,5	148	2,2',3,4,5,6'	192	2,3,3',4,5,5',6
20	2,3,3'	63	2,3,4',5	**PentachloroCB**		149	2,2',3,4',5’6	193	2,3,3',4',5,5',6
21	2,3,4	64	2,3,4',6	107	2,3,3',4',5	150	2,2',3,4',6,6'	**OctachloroCB**	
22	2,3,4'	65	2,3,5,6	108	2,3,3',4,5'	151	2,2',3,5,5',6	194	2,2',3,3',4,4',5,5'
23	2,3,5	66	2,3',4,4'	109	2,3,3',4,6	152	2,2',3,5,6,6	195	2,2',3,3',4,4',5,6
24	2,3,6	67	2,3',4,5	110	2,3,3',4',6	153	2,2',4,4',5,5'	196	2,2',3,3',4,4',5',6
25	2,3',4	68	2,3',4,5'	111	2,3,3',5,5'	154	2,2',4,4,5,6'	197	2,2',3,3',4,4',6,6'
26	2,3',5	69	2,3',4,6	112	2,3,3',5,6	155	2,2',4,4',6,6'	198	2,2',3,3',4,5,5',6
27	2,3',6	70	2,3',4',5	113	2,3,3',5',6	156	2,3,3',4,4',5	199	2,2,3,3',4',5,5',6
28	2,4,4'	71	2,3,4',6	114	2,3,4,4',5	157	2,3,3',4,4',5'	200	2,2',3,3',4,5,6,6'
29	2,4,5	72	2,3',5,5'	115	2,3,4,4',6	158	2,3,3',4,4',6	201	2,2',3,3',4,5',6,6'
30	2,4,6	73	2,3',5',6	116	2,3,4,5,6	159	2,3,3',4,5,5'	202	2,2',3,3',5,5',6,6'
31	2,4',5	74	2,4,4',5	117	2,3,4',5,6	160	2,3,4',4,5,6	203	2,2',3,4,4',5,5',6
32	2,4',6	75	2,4,4',6	118	2,3',4,4',5	161	2,3,3',4,5',6	204	2,2',3,4,4',5,6,6'
33	2',3,4	76	2',3,4,5	119	2,3,4,4',6	162	2,3,3',4',5,5'	205	2,3,3',4,4',5,5',6'
34	2',3,5	77	3,3',4,4'	120	2,3',4,5,5'	163	2,3,3',4',5,6	**NonachloroCB**	
35	3,3',4	78	3,3',4,5	121	2,3',4,5',6	164	2,3,3',4',5',6	206	2,2',3,3',4,4',5,5',6
36	3,3',5	79	3,3',4,5'	122	2',3,3',4,5	165	2,3,3',5,5',6	207	2,2',3,3',4,4',5,6,6'
37	3,4,4'	80	3,3',5,5'	123	2',3,4,4',5	166	2,3,4,4',5,6	208	2,2',3,3',4,5,5',6,6'
38	3,4,5	81	3,4,4',5	124	2',3,4,5,5'	167	2,3',4,4',5,5'	**DecachloroCB**	
39	3,4',5	**PentachloroCB**		125	2',3,4,5,6'	168	2,3',4,4',5',6	209	2,2',3,3',4,4',5,5',6,6'
TetrachloroCB		82	2,2',3,3',4	126	3,3',4,4',5	169	3,3',4,4',5,5'		
40	2,2',3,3'	83	2,2',3,3',5	127	3,3',4,5,5'	**HeptachloroCB**			
41	2,2',3,4	84	2,2',3,3',6			170	2,2',3,3',4,4',5		

Table 3. Occurrence of PCBs in the environment [1].

Environment	Concentration ng/m³	Organism	Contents mg/kg
Air	0.1 - 20	Plankton	0.01 - 20
Water	0.1 – 30 000	Invertebrate	0.01 - 10
Sediments	1.0 - 1000	Fishes	0.01 - 25
		Bird's eggs	0.1 - 500
		Human body	0.1 -10

Another important problem connected with PCBs is their transport in water ecosystems. Water is a carrier of pollutants to plants and animals, via surface runoff to rivers, lakes, and groundwater. The research by Namieśnik et al. [14] demonstrated that transport of PCBs in the river takes place both with the aqueous phase and with suspended matter. Such conclusions were drown on basis of concentration determination in the three phases: water phase, suspension, bottom sediments.

Subsequently, the parameters of PCB distribution in the environment depending on the degree of industralization are very important. The paper by Zalewski et al. [15] presented the comparative analysis of occurrence and concentration of PCBs in bottom sediment samples collected from different types of reservoi's: small and highly urbanized catchments (Sokołówka Reservoir), a large and diversified (agriculture/urban) catchment (Jeziorsko Reservoir) and a small catchment of agricultural impact (Barycz Reservoir). The research proves that concentrations of PCBs in urban areas were highest in comparison with diversified and agricultural catchments. So, the effect of urban catchment on PCB distribution is high.

Living Organisms

The content of PCBs in living organisms is connected with the environment in which they are living. So, concentrations of PCBs in organisms could be correlated with the degree of pollution by these compounds in the environment, by the determination of the bioconcentration factor (BCF, for example in the case of aquatic ecosystems - the ratio of PCBs in biota to PCBs in water) and biota-sediment accumulation factor (BSAF, ratio PCBs in biota to PCBs in sediment). PCBs easily bioaccumulate because of their great affinity for fats.

The concentration of PCBs in freshwater bivalve molluscs (Anodonta complanata) were examined by Ciereszko et al. [16]. These authors found good correlation of mean PCB concentration in mussels with concentration of these compounds in bottom sediments. They estimated BSAF in mussel tissues in the range 3.1 to 5.3 (expected theoretical value equals 2.4). However, BCF obtained were lower than expected by about 30%.

Another organism that was studied in the context of PCB content was pikeperch [17]. The level of PCB concentration in the gonads of pikeperch females from Szczecin, Vistula and Curonian lagoons were studied. The authors have suggested that PCB occurence in pikeperch gonads in different quantitites depends on the region studied.

Pollution of Baltic fish and fishery products by hexachlorobenzene and pentachlorobenzene were studied by Falandysz et al. [18, 19]. Authors indicated the existence of a slight local source of pollution with hexachlorobenzene in the area of the seaport of the city of Gdynia.

As mentioned above, the organisms could be some indicator of pollution in the given area. Zalewski group studied content of PCBs in perirenal fat from red deer obtained from the region of Warmia nad Mazuria in the years 2000-01 [20]. On the basis of the results, authors have concluded that this region is characterized by low levels of pollution caused by PCBs.

Food Products

As mentioned previously, PCBs are very lipophilic, and they tend to accumulate and be decomposed in food chains. In comparison with the other chemical compounds, PCBs have relatively high K_{OW} = octanol/water partition coefficients, in the range from 4.5 for monochloronated biphenyls up to > 8 for higher chlorinated PCBs.

The composition of mixtures of PCBs present in food chains could deviate from the ones found in the environment. In the scientific literature, research on levels of PCBs in foods consumed by the general population or importance of specific food groups to the total intake were conducted [21-22].

Generally, the content of PCBs in food products and human samples (e. g. milk of woman) depend on the region in which the samples were collected. Smoczynski et al. collected samples of human milk from four different regions of Poland, as well as cow's milk and infant formulas [23, 24]. The authors concluded that samples of milk from women living in rural regions contained higher amounts of PCBs, at above 37% in comparison with samples of milk from women living in different regions [24]. Also, the effects of origin of dairy products and their type on the concentration of PCBs were studied by the same group of researchers [23]. The samples of milk, cream, fresh white cheese and butter originating from central, western and southeastern regions of Poland were analyzed. Among samples studied, the significant increase of PCB levels in fresh white cheese were observed. Fortunately, the mean contents of compound analyzed were lower in comparison with permissible standards in Poland.

The determination of PCBs in select components of a food chain (different kind of fodders: for poultry, pigs and cattle) was done by Buszewski et al. [25]. They detected five congeners of PCBs with the detection limit in the range from 85 to 130 µg/kg.

Methods of PCB Determination

Conventional Methods of Determination

During the analytical process, isolation and preconcentration of interest analyte are very important steps. There are many reccomended methodologies for extraction of samples: microwave-assisted extraction (MAE), supercritical fluid extraction (SFE), or accelerated solvent extraction (ASE), just to name a few [26].

The method of isolation of PCBs from matrice is an important step in determining PCBs. Previously, PCBs were extracted from different environmental matrices by means of liquid – liquid extraction (LLE) or Soxhlet extraction. However, nowadays these compounds are extracted by solid-phase extraction (SPE) [27, 13], which is a powerful technique to overcome the drawback of LLE. Moreover, this technique has the property of removing interferents from a sample matrix. The different adsorbent types are used: silica (polar), Florisil (basic), C-8, C-18 (non-polar), and HLB-copolymer of N-vinylpyrrolidone, plus divinylbenzene for cleaning up the samples [28].

Traditional methods such as Soxhlet extraction need a big volume of solvents and a long extraction time, then the new extraction techniques are required. Analytical procedure was validated by Lulek et al.: reduction of the amount of organic solvent from 300 to 60 ml, and shortening of extraction time from 24 h to 250 minutes, in comparison with the Soxhlet technique. The procedure consisted of extraction and clean-up of extracts with concentrated sulphuric acids and solid phase extraction (SPE) on Florisil [13].

In the beginning, the PCBs were determined by packed column gas chromatography (GC) in combination with detection by mass spectrometry (MS) [26]. During the last few years, analytical methods have become more sophisticated and nowadays, cappilary (high resolution) GC with high resolution MS [HRGC-HRMS] are routinely used in analytical laboratories [13, 27].

Alternative Methods of Determination

Conventional analytical methods of detection of PCBs could be supplemented by immunochemical methods (immunoassays) [29]. These methods are rapid, sensitive and cost-effective, and combine chemistry and immunology. The working principle of this technique relies on interaction between an antibody (analytical reagent) and an antigen (target analytes).

Another method, which could be applied for determinatation of PCBs or products of their degradation, is potentiometry. Originally, this technique was used for determination of ionic species. Recently, the determination of uncharged molecules using this technique is disseminated [30-32]. The main element of measuring system is electrode with liquid membrane type, which contains a lipophilic receptor capable of interacting with analytes present in the aqueous phase. The results of recognition of chloro-derivatives of phenols and discrimination between *ortho*-, *meta*- and *para*-isomers of chlorophenol was presented by Piotrowski et al. [33]. The sensitivity and selectivity of such sensors depends on acidity and liphilicity of isomers of chlorophenol as analytes.

Evidently, the alternative methods cannot replay the conventional ones, but they could be helpful (at least for screaning purposes).

Toxicity of PCBs

It is well known that PCBs are toxic to endocrine, immune and nervous systems, especially in the early developmental stages, and could cause adverse effects on reproduction and development of animals and humans [34]. So, for example, the consumptions of marine food by mothers during pregnancy and breast feeding could affect neurological or neurobehavioral disorders of children.

Some papers have informed that PCB and MeHg can act synergistically to influence the nervous system. The papers by Świercz et al. [35, 36] show the influence of PCBs on the nervous systems in rats. The authors' aims were to asses the influence of separate or combined perinatal exposure to MeHg and PCB 153 on brain catecholamine content in maturity. They summed up the results that gender may be important determinants of rats' vulnerability to PCB 153 [35]. Moreover, perinatal exposure to MeHg and/or PCB, separated or combined, may cause some changes in catecholamine levels in a mature brain. However, neither synergistic nor antagonistic activity of these substances was confirmed.

The same authors also studied how perinatal exposure of PCB 153 determines behavioural sensitivity and sensitizability to the psychostimulant amphetamine (AMPH) in adulthood [36].

Degradation of PCBs

PCBs belong to compounds strongly resistant to destruction, because of their high chemical stability. So, determining the methodology of their degradation is a great challenge.

The newest trend in destroying PCB waste is application of non-combustion technologies (e. g. chemical treatment) instead of incineration or high temperature pyrolysis. Then, many investigations have been conducted in order to improve chemical methods. Among them, dechlorination [37] and photocatalytic degradation [38] are examples of degradation treatment, to name just a few. Biodegradation of PCBs could not also be omitted. Microbial degradation by PCB – degrading bacteria has been described and these bacteria are isolated and characterized [39].

Degradation of PCBs by Fenton`s reaction has been applied successfully by Polish scientists [40]. In this reaction, the mixture of hydrogen peroxide nad Fe^{2+} produce hydroxyl radicals, which are characterized by high standard reduction potential in acid media, which in consequence causes mineralization of organic compounds to CO_2, H_2O and inorganic ions. Selected congeners of PCBs (PCB28, PCB52, PCB101, and PCB138) present in aqueous solutions at ppb concentrations were degradated by Fenton's reagent by Prządo et al. [10]. Within the first 24 hours of experiments, the concentration of two congeners, PCB28 and PCB52, decreased rapidly. On the other hand, the other two, PCB101, and PCB138, were poorly degraded. During the process, the concentration of chlorine ions slightly increased, which indicated a lack of mineralization of substrates.

Conclusions

1. The presence of polichlorinated biphenyls was identified in almost every environmental sample, starting from aquatic systems to mother`s milk. Their concentration depends on the nature and location of the particular environmental samples.
2. Evaluation and validation of determination methods of selected PCB congeners and their mixtures is still a challenge for analytical chemists.
3. Because of high toxicity of these compunds to humans and animals, their content should be monitiored in food products, especially in fish, fats, eggs and meat.
4. Studies on efficient degradation process of PCBs are still needed, particularly because these compunds are extremely resistant to chemical, photochemical and biological degradation.

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